Determination of the Half-life of ³⁷Ar by Mass Spectrometry

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³⁷Ar is produced in nature by cosmic-ray spallation and by neutron capture processes, and decays by electron capture to ³⁷Cl. The half-life of isotope is approximately 35 Knowledge of the half-life is important to ⁴⁰Ar/³⁹Ar dating¹ and various problems in cosmochemistry including neutrino detection2. Previous determinations³⁻⁸ of the ³⁷Ar half-life have been based on activity measurements. Here we report mass spectrometric data monitoring the ratio of 37Ar to stable 36Ar as a function of time after neutron activation9. These data provide an independent corroboration of the activity-based results, and yield a more precise value for the half-life than all but the most recent of those experiments.

Optical grade fluorite (CaF2) was irradiated in 11 distinct neutron irradiation runs of 7 to 100 hours duration at the 1 MW TRIGA reactor at Oregon State University. 37Ar and 36Ar are produced by fast neutrons via the ${}^{40}\text{Ca}(n,\alpha){}^{37}\text{Ar}$ 40 Ca(n,n α) 36 Ar reactions, respectively. Normalizing radioactive ³⁷Ar to stable ³⁶Ar enables using decay of the 37Ar/36Ar ratio to constrain the ³⁷Ar half-life. Because atmospheric ³⁶Ar is present in variable concentrations, a correction using ⁴⁰Ar is required. This correction, which assumes that all ⁴⁰Ar is of atmospheric origin, yields the reactor-produced ³⁶Ar (³⁶Ar_{Ca}). Analysis of unirradiated samples of the same fluorite confirms an atmospheric 40Ar/36Ar ratio. ³⁶Ar_{Ca} comprises 79-99% of all ³⁶Ar in the irradiated fluorite samples analyzed, and the atmospheric correction introduces small errors compared with those from mass spectrometry. For each analysis, 0.2 to 3 mg of irradiated fluorite were degassed in ultrahigh vacuum with either an argon-ion or a Nd-YAG laser, and purified gas was analyzed with a noble gas mass spectrometer. The amount of fluorite analyzed was adjusted to maintain ³⁷Ar yields at least 50 times background. The precision of individual $^{37}Ar/^{36}Ar$ measurements ranges from $\pm 2\%$ to $\pm 35\%$, with a median of $\pm 11\%$.

Argon ion beam currents were measured on a single electron multiplier using magnetic field switching to cycle sequentially between argon isotopes. All data reported herein are corrected for background (measured between every 1-3 samples) and mass discrimination (1.00282 ± 0.00215 to 1.01133 ± 0.00200 per amu) based on average values of $^{40}\text{Ar}/^{36}\text{Ar}$ from air pipettes interspersed with unknowns.

Regression analysis of these data yields a value of 34.95 ± 0.08 days, which is at least twice as precise as all but the most recent of the previous activity-based measurements. The utility of methods involving mass spectrometry for determining decay constants has been exploited for several decades by earth scientists. To our knowledge, such experiments have always used ingrowth of a daughter nuclide rather than dimunition of the parent. In difficult cases, including that of 37 Ar which involves experimentally challenging detection of soft x-rays and/or Auger electrons, it would seem desirable for nuclear data evaluations such as ENSDF or NUBASE to expand their literature searches and take account of all relevant data.

Footnotes and References

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